Integrating *in situ* X-ray tomography and computational analysis to investigate buried electrode/electrolyte interfaces in solid-state lithium batteries

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As the need for a clean and viable environment is growing, the development of energy storage systems is becoming increasingly important. Among the various energy storage systems, solid-state batteries using lithium metal has become an attractive option due to its potential to improve safety and energy density. Therefore, this technology can essentially be used in electric vehicles, consumer electronics, and other applications where reliable, long-lasting energy storage is needed [1,2]. However, the potential of lithium metal batteries is still not fully explored due to their intrinsic complexity: the structure, components, and chemistry of lithium metal batteries are highly correlated, making it difficult to gain in-depth understanding. Moreover, the interfaces, which are critical to the performance of solid-state lithium batteries, are buried throughout the battery stack and therefore inaccessible to conventional analysis techniques. The recent development of advanced 3D X-ray computed tomography (XCT) holds the promise to offer unprecedented insight into the internal operation of lithium metal batteries, as the technique enables non-invasive *in situ* and *in operando* studies [3]. XCT is not only helpful in extracting hidden morphological information of the battery but also can be utilised in combination with numerical modelling. The present research employs *in situ* XCT to investigate the buried lithium/solid polymer electrolyte interface while performing a galvanostatic test at different current densities in a purpose-built cell setup with optimized cell stack pressure. The dedicated cell setup, which is highly compatible with the micro-XCT imaging and electrochemical testing, successfully provides highly-resolved and high signal-to-noise 3D tomographic datasets at different stages of cycling. The datasets were filtered and subsequently segmented into morphologically distinct phases. In parallel, a digital twin of the cell was subjected to the same galvanostatic cycling protocol. By fusing the data from model and experiment, we obtain a unique view on the microstructural changes along the buried interfaces during cycling, the locally produced heat by the electrochemical reactions, the volume changes of each phase, and the local stress state induced by differential volumetric changes. These results are also supported by post-cycling examination with techniques as XPS to get detailed chemical information. We believe that the proposed approach will help to gain in-depth understanding of interfacial changes over prolonged cell cycling. By gaining deep understanding of the structure and chemistry aging of these lithium metal batteries, we believe it is possible to improve them and thus to make a significant contribution towards sustainable energy solutions.

References:

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